Introduction

Persistent organic pollutants (POPs) are now nearly ubiquitous in their distribution over the Earth. They can be found in remote locations distant from industrial and agricultural regions, as well as close to their point of introduction into the environment. Their propensity for long-range environmental transport, and the extent of their distribution, are evident from the levels reported for the Great Lakes, Alaskan Arctic, and marine ecosystems. This chapter provides information on how POPs are transported such long distances by winds, ocean currents, and migratory animals. The focus is on transport of POPs into the United States, recognizing that POPs also leave the United States and have impacts on other countries through similar processes. Particular attention is directed to transport in the atmosphere, because this is the principal medium through which POPs are distributed globally, either as vapor or on particles. Examples are provided of the transport of pollutants between continents, accompanied by an overview of the atmospheric chemistry of POPs relating to entrainment, degradation, and deposition. Methods for tracking the transport of POPs through the atmosphere are discussed, with examples provided of the pathways they may take. This discussion is followed by a summary of information on hydrologic movement and transport of POPs via migratory species.

The behavior of POPs in the environment is complex because they are multimedia chemicals, existing and exchanging among different compartments of the environment such as the atmosphere, natural waterbodies, soil, and sediments, where they degrade at different rates over time. POPs are also referred to as semivolatile, meaning they can be present in more than one phase in the atmosphere, either as gases or attached to airborne particles. The fate and preferential transport of a POP are strongly determined by its specific physical chemistry properties, even its particular isomer (i.e., same molecular formula but different spatial structure). The different affinities of POPs for soil particles, water, and/or lipid molecules, and their rate of volatilization will determine the pathway each species or isomer is likely to take in its journey through the environment. These properties also influence how far and fast a POP can move from where it was released into the environment.

Comprehensive monitoring studies support the conclusion that POPs concentrations are generally highest in areas where they were once released, or are still being released. Concentrations generally decrease with increasing distance from such source areas. Thus, concentrations are most strongly dependent on past and present release rates in the immediate vicinity under investigation, and are also strongly influenced by regional releases (Kalantzi et al., 2001). In addition, however, the finding of surprisingly high concentrations of POPs in polar areas, particularly the Arctic (Bidleman et al., 1989; Barrie et al., 1992; Iwata et al., 1993), has led to an emphasis on the natural processes that are responsible for this wide-ranging transport through the global environment. These processes include:

✳ Volatilization of POPs from terrestrial and/or aquatic surfaces into the atmosphere

✳ Adsorption of POPs vapor onto particles already entrained in the atmosphere

✳ Entrainment from a surface of particles with an adsorbed POPs layer into the atmosphere

✳ Transport of air masses throughout a hemisphere by means of persistent, large-scale circu-

lation patterns or by means of episodic rapid transport processes

✳ Photochemistry and interaction of POPs with free radicals, which can modify and degrade their chemical form while they are undergoing atmospheric transport

✳ Deposition by means of wet (e.g., snow, rain, mist) or dry (e.g., turbulent transport and particle settling) atmospheric processes onto terrestrial and aquatic surfaces of POPs that are either in vapor form or adsorbed onto particles

✳ Transport of POPs in aquatic systems by means of flowing water (primary surface flows such as rivers and ocean currents)

✳ Transport of POPs in terrestrial and aquatic ecosystems in the lipids of migrating mammals, fish, and birds

✳ Deposition of POPs into aquatic sediments

✳ Eventual physical accumulation in receptor locations, uptake, and bioaccumulation to reconstitute concentrations in ecosystems and humans

Atmospheric Transport of Pollutants to the United States

When describing winds in the atmosphere and their ability to transport trace substances over long distances (e.g., the equator to the pole or around the globe) a convenient first approximation is to focus on their long-term average behavior. This focus is useful because winds fluctuate in strength and direction in passing cyclonic and anticyclonic weather systems, resulting in net displacements that are relatively small compared with the global scale. Average, or prevailing, winds blow much more in east-west directions than in north-south directions. In midlatitudes, the prevailing winds are westerly (i.e., from the west), whereas easterly winds prevail at very high latitudes and in the subtropics (e.g., southern Florida), the latter especially in spring and summer. In principle, pollutants released at the surface can travel eastward completely around the globe at midlatitudes in about 10 days if they are lifted by convective activity (strong upward air motions that produce clouds) to the

altitude of the jet stream. The average wind speeds at this level are much stronger than they are near the surface and can exceed 100 mph in the core of the jet stream (e.g., Stull, 2000), and the wind speeds and directions are more constant than they are near the surface. In contrast to this rapid west-east transport in the jet stream, it takes several months for pollutants to spread from equator to pole. It can take even longer, about a year to a year and a half, for pollutants to cross the equator and be evenly distributed over the globe (Warneck, 1988). Mixing caused by small-scale turbulence normally causes pollutants to disperse so they no longer form a coherent plume that can be tracked over long distances. However, on occasion, plumes of airborne substances remain coherent and can be tracked for long distances.
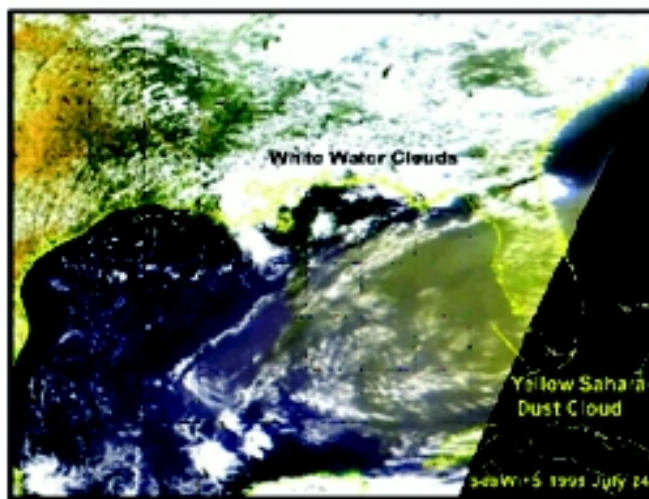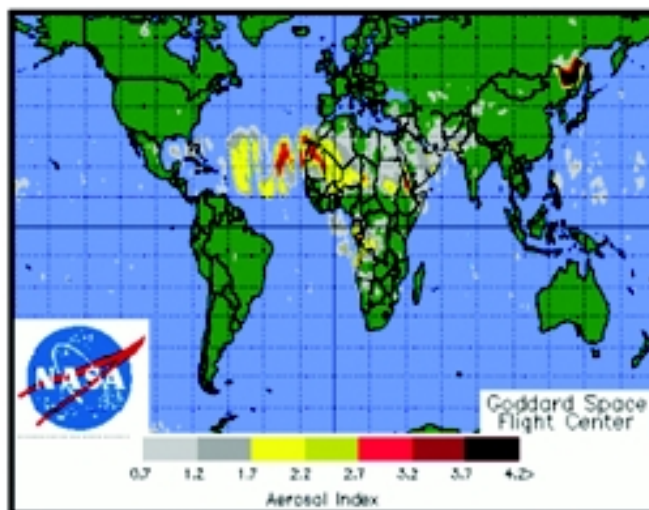

Figure 7-1. Atmospheric transport of Saharan dust to the United States.

Source: National Aeronautics and Space Administration (2001).

Much of the evidence for long-range transport of airborne gaseous and particulate substances to the United States focuses on dust or smoke because these are visible in satellite images or when deposited. Examples include transport of dust from the Sahel and Sahara Desert in northern Africa, dust from the Taklamakan and Gobi Deserts in Asia, and emissions from uncontrolled wildfires in Central America and southern Mexico. Windblown dust from individual dust storms in the Sahara Desert has been observed in satellite images as plumes crossing the Atlantic Ocean and reaching the southeast coast of the United States. These storms can last for several days at a time. A false color satellite image obtained by NASA's Earth Probe TOMS satellite showing successive pulses of Saharan dust propagating eastward across the Atlantic Ocean and reaching Miami is presented in the top portion of Figure 7-1. The bottom portion of Figure 7-1 shows a true color image of the dust cloud over Florida obtained by NASA's Sea Star satellite on the same day. Analysis of data obtained by the IMPROVE (Interagency Monitoring of Protected Visual Environments) network indicates that incursions of Saharan dust into the continental United States have occurred, on average, about three times per year. These events have persisted for about 10 days, principally during the summer. As might be expected, the frequency of Saharan dust events is highest in the southeastern United States. About half are observed only within the State of Florida, and these are associated with dense hazes in Miami (see Figure 7-2) such that African dust is the dominant aerosol constituent in southern Florida during the summer (Prospero, 1999a). Puerto Rico and the Virgin Islands are even more strongly affected, as might be expected. Figure 7-3 shows a false color satellite image of the passage of a cloud of dust across the Pacific Ocean to North America. This dust cloud was raised by a storm in the Gobi and Taklamakan Deserts in April 2001. The highest concentrations of Asian dust can be seen over the Aleutian Islands. Also shown in Figure 7-3 is a dust cloud from northern Africa traveling eastward over the Atlantic Ocean.

Biomass burning for agricultural purposes occurs normally during the spring of each year in Central America and southern Mexico. During the spring
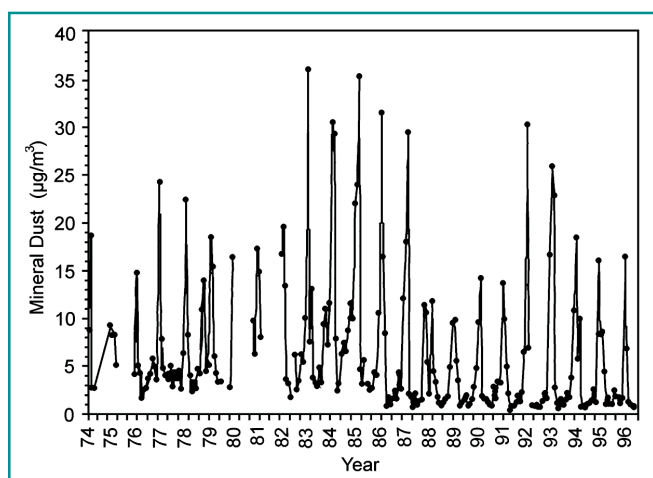

Figure 7-2. Saharan dust episode chronology impacting Miami, FL.
Source: Prospero (1999a).

of 1998, fires burned uncontrollably because of abnormally hot and dry conditions associated with the intense El Niño conditions in 1997–1998. Figure 7-4 shows the extent of the spread of the particles emitted by the fires. Concentrations of particles throughout the central and southeastern United States were elevated substantially, such that National Ambient Air Quality Standards for particulate matter were exceeded briefly in St. Louis, MO, and in a number of other cities in the central United States. The reader should bear in mind that the atmospheric processes that transport dust or smoke, as given in the examples above, are also transporting pollutants, including POPs, either as
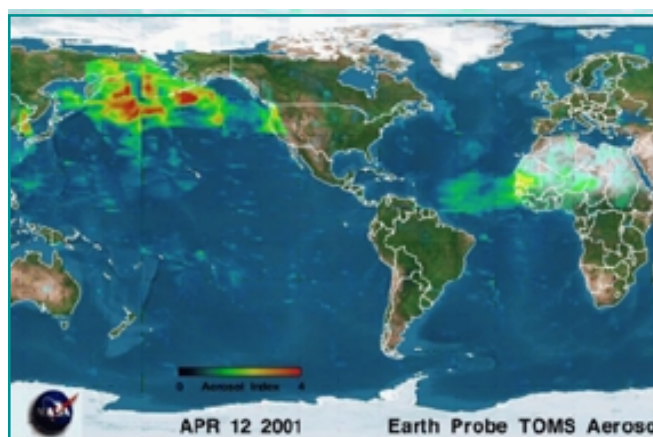

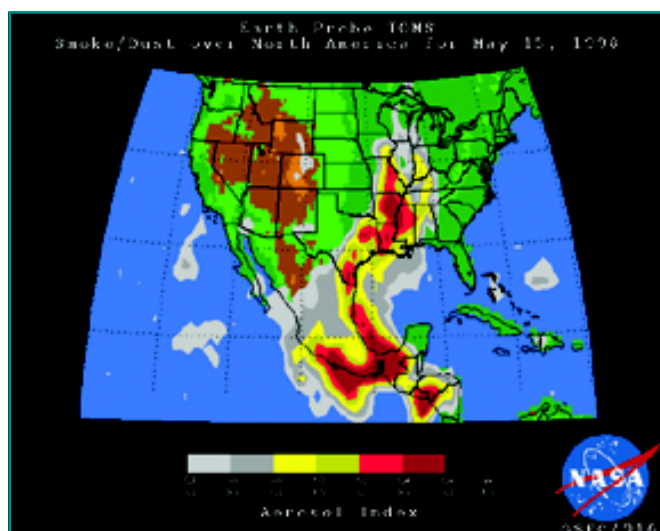*Figure 7-3. Asian dust storm episode crossing the Pacific Ocean. NASA.*

Figure 7-4. Smoke from Central American fires. NASA.

vapor or attached to particles. Indeed, micro-organisims including various fungi and bacteria have been found attached to North African dust particles in Caribbean air samples (Griffin et al., 2001). Pollutants, including trace metals that may have been emitted in North Africa or Europe, have been found along with North African dust particles in Miami, FL (Prospero, 1999b).

Rapid transport of pollutants from south to north can also occur, bringing pollutants from mid-latitudes to the Arctic in a few days. Many of these pollutants are relatively short-lived with respect to degradation in the atmosphere, and are unlikely to reach the Arctic by the average winds described above. These transport events occur during winter and are caused by transient weather events that result in strong winds directed toward the north in a narrow current lying typically between a strong high-pressure region to the east and a low-pressure system to the west. These weather situations occur mainly in the former Soviet Union, although they can also occur in North America. These events result in the formation of Arctic haze, which regu-larly affects air quality in Alaska (Shaw and Khalil, 1989).

Although these episodes are infrequent, the amount of POPs deposited can be substantial, given the otherwise pristine nature of some of the receptor locations. For example, Welch et al.

(1991) documented a long-range transport event that deposited thousands of tonnes of Asian dust onto a region of the central Canadian Arctic over a brief (<3-day) period. Analysis of the resulting brown snow revealed elevated levels of the follow-ing POPs: PCBs (6.9 ng/g particles), DDT (4.2 ng/g), toxaphene (3.0 ng/g), HCB (0.7 ng/g), chlordane (0.6 ng/g), heptachlor, and dieldrin, along with other organic pollutants such as polycy-clic aromatic hydrocarbons (PAHs) and hexa-chlorocyclohexanes (e.g., lindane). The authors estimated that this single episode may have contrib-uted up to 10% of the annual loading of DDT to lakes in this region, and up to 1%–3% for the other measured POPs (depending on loading scenario assumptions).

Atmospheric Chemistry of POPs

Many POPs, including pesticides and PCBs, are classified as semivolatile, meaning that they can exist either as gases or attached to particles. The relative amounts of these forms depend on air temperature. Less volatile POPs tend to partition into surface reservoirs such as soil, vegetation, rivers, and oceans, where they associate with or-ganic matter. Warmer temperatures favor their evaporation and residence in the atmosphere as gases; colder temperatures favor their deposition to the Earth's surface and their incorporation into airborne particles. Generally, warmer tempera-tures are found close to the Earth's surface with decreasing latitude (tropics), whereas colder tem-peratures are found with increasing latitude (polar) or altitude. Characteristic temperatures at which half of a particular compound ($T_{1/2}$) would be present as a gas can be calculated to indicate the tendency of a POP to remain as a gas or to attach to a particle (Wania et al., 1998). The more vola-tile POPs, such as hexachlorobenzene (HCB), reach this temperature at −36°C; dieldrin at −11°C; DDE −2°C; DDD +7°C; DDT +13°C; and PCBs with at least 6 Cl atoms at about +10°C. As the number of Cl atoms increases in PCBs, polychlori-nated dibenzo-*p*-dioxins (PCDDs), and furans (PCDFs), this characteristic temperature also in-creases, such that molecules with more than 5 Cl atoms tend to be partitioned mainly onto particles (Brubaker and Hites, 1997). Compounds such as

mirex and toxaphene are expected to be found mainly attached to particles, because they have very high values of $T_{1/2}$. Observations show that compounds with the lowest $T_{1/2}$ tend to have the highest potential for global scale transport and to remain in the gas phase even at high latitudes, whereas compounds with the highest values of $T_{1/2}$ tend to remain concentrated close to their sources.

Although the meteorological processes transporting gases and particles are the same, particles and gases are subject to different removal processes that affect the time they remain airborne. Soluble gases can be removed from the lower atmosphere by being incorporated into cloud droplets that then fall out as rain, or by being washed out by falling raindrops. This limits their lifetime in the atmosphere typically to several days, and also limits the distances they can travel from their sources. However, gas-phase POPs are not very soluble and so their removal by precipitation is not effective. For example, the removal time for chlordane, dieldrin, and PCBs can be up to several years by this mechanism (Atlas and Giam, 1981).

POPs present as atmospheric gases can be destroyed by atmospheric photochemical reactions involving hydroxyl (OH) radicals in the atmosphere. Although data are sparse, reactions involving other species or photodegradation by solar ultraviolet radiation are considered to be minor loss processes. Calculated atmospheric half lives, $t_{1/2}$ (defined as the time it takes to reduce their concentrations by one-half due to reaction with OH radicals) for several compounds are shown in Table 7-1, along with their concentrations in both the gas and particle phases observed on the shores of the Great Lakes. As can be seen, $t_{1/2}$ in the atmosphere ranges from a couple of days for DDT to a couple of years for HCB. Notably, the presence of hydroxyl radicals is integrally linked to the presence and level of sunlight. Absence of sunlight in polar regions for many months of the year effectively eliminates the generation of hydroxyl radicals, thereby greatly increasing the persistence of atmospheric pollutants that would otherwise be degraded through this mechanism during daylight.

If POPs become attached to particles, their lifetimes in the atmosphere are determined by particle removal mechanisms, in addition to reactions involving hydroxyl radicals or other free radical species in the particles and photodegradation by solar ultraviolet radiation. However, the effectiveness of these processes in particles has not been studied as well as for the gas phase. The lifetime of particles in the lower atmosphere is about 1 week with respect to removal by precipitation. However, particles above cloud layers can remain airborne much longer and hence can be transported over much longer distances. Particles also can be deposited on the Earth's surface when they are transported downward by turbulent air motions (dry deposition). Atmospheric lifetimes with respect to dry deposition depend on particle size, meteorological variables near the surface, and microphysical conditions at the air-surface interface. Lifetimes in the atmosphere of fine particles (i.e., smaller than a few micrometers in diameter) with respect to dry deposition are about a couple of weeks (U.S. EPA, 1996).

Deposition on to the surface does not, however, mean that a POP has been destroyed. Through a variety of mechanisms, such as microbial activity and photochemical reactions occurring near the

Table 7-1. Atmospheric concentrations of selected POPs at IADN[a] sites and their estimated globally averaged atmospheric half-lives ($t_{1/2}$)

Compound	Concentration Range (pg/m³)[b]		$t_{1/2}$[c]
	Gas	**Particle**	
Hexachlorobenzene	80-130	0.1-0.2	728 d
Dieldrin	14-34	1.5-3.2	6.2 d
PCB44	3.4-14	0.09-0.2	16 d
DDT	3.9-91	0.3-3.6	2.3 d

[a] Integrated Atmospheric Deposition Network. IADN consists of five sites situated on each of the Great Lakes.
[b] 1 pg = 1 trillionth (10^{-12}) of a gram.
[c] Defined here as the time needed in days to reduce the concentration of the compound by one-half, using a globally averaged OH concentration of 1.0×10^6 OH/cm³ (e.g., Krol et al., 1998) and a mass weighted mean atmospheric temperature of 273°K (0°C).

Source: Hoff et al. (1996) for concentration data.

surface of the soil or a waterbody, POPs that were attached to particles may be released and evaporate into the atmosphere. Only through processes such as reaction with OH radicals in the atmosphere, or biologically mediated reactions in either soil or natural waterbodies, is a POP finally gone from the environment. These processes are all strongly temperature dependent and proceed faster at higher temperatures, so that POPs tend to persist longer at higher altitudes or latitudes. Lifetimes of most POPs due to degradation in the marine or terrestrial environment are estimated to be several years, with degradation occurring more rapidly in tropical than in polar regions. A degradation product of the original POP may also be a POP, or may be toxic in the environment. For example, dieldrin is formed during the degradation of aldrin.

Global Distillation of POPs

An intriguing consequence of the combined semivolatile and persistent nature of POPs is their potential to volatilize in warm regions, be deposited in colder ones, and repeat this process until a location is reached where it is too cold for the POP to again revolatilize. This process is similar to industrial distillation processes, such as separating petroleum products from crude oil. On a planetary scale, global distillation has been hypothesized to result in a net transport of POPs from lower latitudes to high latitudes (polar regions) in a series of jumps (Wania and Mackay, 1996) (Figure 7-5). Because of the normal decrease of temperature with increasing latitude, compounds will tend to condense on surfaces as they are transported northward by winds associated with passing weather systems. Various thermodynamic constants can be used to estimate the "stickiness" of POPs to a surface. One of the most useful is the octanol-air partition coefficient (K_{OA}), which is measurable in the laboratory and relates to the tendency of a POP to adhere to organic matter in the soil or natural waterbodies. Values of K_{OA} range over many orders of magnitude, and the higher its value the greater the tendency for a POP to associate with organic matter.

The environmental levels of POPs predicted in remote locations following global distillation de-

pend heavily on the physicochemical properties of the particular POP and the meteorological factors generating poleward transport. The balance between POPs concentrations in temperate regions versus their preferential accumulation toward the poles is influenced by the following factors:

✳ *Source proximity:* The strength and proximity of emission sources act as the primary forces generating pollutant levels. All things being equal, pollutant concentrations should be greater at the source and taper off with distance, with this tapering effect proportional for all pollutants. But a variety of factors can differentially influence the transport potential of POPs and their isomers, leading to differential accumulation.

✳ *Physicochemical properties of different POPs:* As already noted, differences among POPs regarding their propensity to exist in the vapor phase (e.g., K_{OA}, Henry's law constant, vapor pressure) have an impact on each POP's susceptibility to global distillation and the rate at which such movement can occur. Differences occur even within families of POPs, such as among the 209 PCB congeners. Lower chlorinated PCBs, with two or three chlorines, are more volatile and amenable to global transport. They tend, however, to be less persistent than the higher chlorinated PCB congeners, somewhat countering their propensity to preferentially move toward the poles.
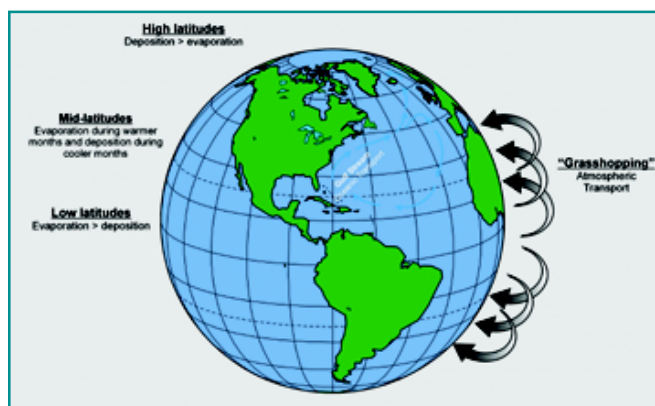

Figure 7-5. Grasshopper effect moving POPs poleward.
Source: Adapted from Wania and Mackay (1996).

✳ *Temperature dependence of physicochemical properties:* Decreased temperatures and reduced microbial activity toward the poles increase POPs' persistence, thereby inherently increasing the potential for physical accumulation (see Chapter 9). Decreased temperatures also alter environmental media partitioning properties, such as the water-air partition coefficient (Henry's law constant), increasing the proportion of a chemical in water compared with the amount in the adjacent air column. Hexachlorocyclohexane (HCH) isomers change from an aquatic persistence of only a few days in the tropics to many months or years in Arctic waters (SETAC, 2000). The Henry's law constant also changes for the HCHs, increasing the proportion in water compared with air as the environment cools. Both of these changes contribute to the higher absolute levels of HCH in Arctic waters compared with those that had been found close to the presumed sources in Asia (Iwata et al., 1993).

Temperature-dependent gradients have been demonstrated on a global or regional scale for several of the more volatile POPs, although direct evidence for transfer of POPs from the tropics to the Arctic remains to be demonstrated (SETAC, 2000). Calamari et al. (1991) reported that HCB concentrations in vegetation increased with latitude by almost a factor of 10 between the tropics and polar regions, whereas concentrations of the less volatile DDT were found to be higher in tropical compared with polar areas by almost a factor of 100. Simonich and Hites (1995) reported similar results on the global distribution of 22 organochlorine compounds in tree bark samples. Here, the distribution of relatively volatile organochlorines, such as HCB, was dependent on latitude, whereas less volatile and persistent organochlorines (e.g., endosulfan) remained close to the region of release. Iwata et al. (1993) measured geographic gradients for HCH in sea water on a global scale, with higher concentrations found toward the poles even though emission sources were in temperate and tropical regions, predominantly Asia. Sea water and air concentrations of DDT were substantially higher near the sources measured in Asia.

Increased altitude and the related decrease in temperature also are associated with increasing levels of POPs, as demonstrated over a 770 to 3,100 m altitude gradient in the western Canadian Rocky Mountains (Blais et al., 1998). Increasing snowfall at higher altitudes led to a 10-fold increase in deposition with height of less volatile compounds, such as DDT. For the more volatile organochlorines (e.g., lower chlorinated PCBs, HCH, and heptachlor epoxide) this increase with altitude was up to 100-fold, demonstrating enhancement through cold-condensation effects. The concentration gradient with altitude is particularly informative because it was performed in a single geographic location, making the study less likely to be confounded by proximity to sources, which can occur when there are several geographically dispersed measurement sites.

Global-scale distillation effects were observed originally for the heavy isotopes (deuterium, oxygen-18) compared with the light isotopes (hydrogen and oxygen-16) of water vapor (Dansgaard, 1953). Light isotopes are more volatile than their heavier counterparts. The ratio of light to heavy isotopes increases with latitude, because of selective condensation of the heavier isotopes in accord with the Rayleigh distillation formula. For POPs, current field data relate to a small subset of the most volatile persistent organic substances, consistent with theoretical predictions (Wania and Mackay, 1996), i.e., hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), hexachlorocyclohexanes (HCH), and, to a lesser extent, chlordane.

The basis of empirical support for global distillation comes from the differential increase in accumulation of more volatile PCB congeners (lower chlorinated) and HCH isomers ($\alpha > \gamma$) in higher latitudes. Many of the factors noted above as influencing POPs levels in remote locations can be seen affecting, and are consistent with, these measured congener and isomer ratios. Total PCB levels were found to decline with increasing latitude (consistent with increasing distance from the source), but the levels of di- and trichlorobiphenyl stayed relatively constant with latitude (they are more volatile), thereby increasing as a proportion of the total PCB

mix (Muir et al., 1996; Ockenden et al., 1998a). Notable, too, is the delayed onset of PCB deposition to sediments in high Arctic lakes compared with midlatitude and subArctic lakes, providing further evidence of ongoing global distillation (Muir et al., 1996). Similar latitude-related ratio changes favoring more volatile compounds in northern, colder sites were reported for α-HCH versus γ-HCH (lindane), the former having a higher Henry's law constant and vapor pressure (Ockenden et al., 1998b).

Calculating and Modeling Atmospheric Transport of POPs

The distribution and movement of POPs in the atmosphere can be calculated by computer modeling. The outputs from these models may be used for making predictions of future trends, for evaluating the effects of control strategies, and for improving understanding of the processes controlling the distribution of POPs. A complete model of the distribution, ultimate fate, and trends for POPs in the environment would include modules calculating the changes of emissions with time; transport by the atmosphere and oceans; deposition to the surface and volatilization from the surface; transfers to rain and snow; and degradation in the atmosphere, oceans, terrestrial waters, and soils. Although information and models are available for each of these modules for some POPs, a complete and validated global model has not been published, principally because of the complexity of the calculations and the uncertainties in input parameters. More pragmatically, because of their persistence, POPs concentrations can be directly measured in the animal species of interest, reducing the need for modeling to determine dose and risk. To date, POPs models have focused on either (1) bulk transfers among different media (i.e., multimedia models) or (2) simulations of atmospheric movement. The multimedia models currently include simplified treatments of transport, whereas the atmospheric models include highly simplified treatments of the transfers among the different reservoirs. These two modeling fields are now merging. Multimedia models are addressed in Chapter 9 of this report (see also SETAC, 2000). The remainder of this section focuses on air transport modeling.

There are two basic approaches for calculating the transport of pollutants in the atmosphere. The first approach is to calculate trajectories, which are the three-dimensional paths followed by the center of mass of an imaginary air parcel, either forward from a source or backward from a receptor location. These are known as Lagrangian models. The second approach is based on numerical grid models, which include a number of physical processes such as the mixing of air parcels, chemical transformations, emissions, and deposition to the surface. These calculations are performed on individual boxes in a three-dimensional grid matrix, thereby tracking the dispersion of a pollutant across this grid (Eulerian models). Both approaches rely on the use of meteorological data obtained from ground-based measurements, weather balloons, and satellites. In some cases, global climatological model simulations are used rather than real observations.

Both approaches have advantages and limitations. The first, or trajectory, approach has the advantage of simplicity, although it minimizes the physical processes mentioned above. For example, after a short time, typically a few days, the air parcels lose their identity because of mixing. On the other
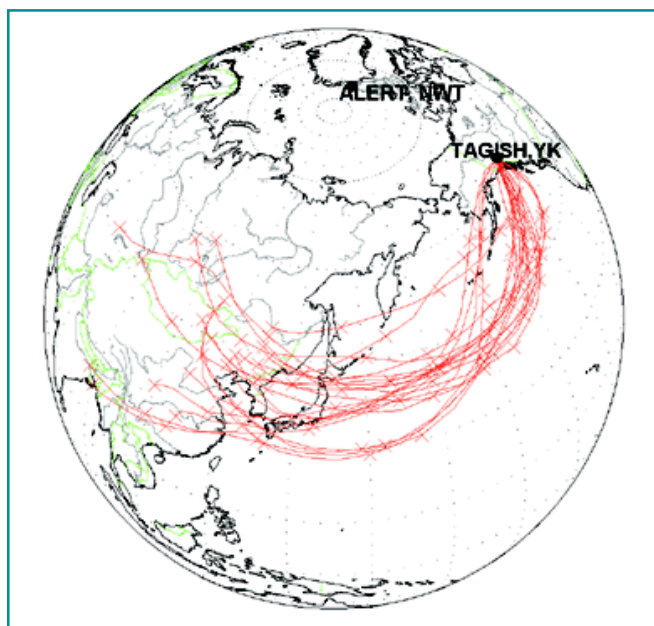

Figure 7-6. Air movement back trajectories from Tagish, Yukon, to Asia.

Source: Bailey et al. (2000).

hand, although the second, or dispersion modeling, approach includes these important processes, they are subject to considerable uncertainties. NOAA's Air Resources Laboratory maintains a Web site (*http://www.arl.noaa.gov/ready.html*) for researchers on which it is possible to calculate trajectories extending either backward or forward for anywhere in either the Northern or Southern Hemispheres.

Back trajectory models are particularly useful to ascertain the source of pollutants. For instance, high levels of a number of organochlorines, such as α-HCH, γ-HCH, DDT, and chlordane, were found in snow samples at a monitoring site in Tagish, Yukon, Canada (Bailey et al., 1999). As shown by the trajectories in Figure 7-6, the pollutants were associated with long-range transport from Asia, occurring generally within the previous 5 days. Coupling emissions data with the trajectory methods can help determine the probability that POPs observed at monitoring sites came from particular source regions. For example, results of this approach suggest that many PCBs and DDT/DDE seen at the Lake Ontario IADN monitoring site may have come from the southeastern United States (Song et al., 2001).

The results of the calculation of a large number of backward trajectories can be assembled to trace "transport pathways" leading to a particular monitoring site. Several thousand trajectories from Alaska were calculated backward for 10 days during the entire year of 1999. Results for 4 months of the year (January, April, July, and October) are shown in Figure 7-7 to give an idea of the seasonal variation in the transport pathways through the atmosphere (Husar and Schichtel, 2001). The different-colored shading in Figure 7-7 reflects the probability that trajectories passed over a given area before arriving at the Alaska Peninsula National Wildlife Refuge in the Aleutian Islands. The boundaries of each shaded region represent lines of constant probability. The areas shaded in red have the highest probability of being traversed by trajectories, whereas those shaded in light blue have a lower probability. Similar calculations for a number of receptor sites throughout the United States are shown in Appendix A. By comparing the locations of the dust and smoke plumes shown in Figures 7-1, 7-3, and 7-4 to the transport pathways shown in the figures in Appendix A, it can be seen that the plumes visible in the satellite images occur within defined transport pathways. Even though
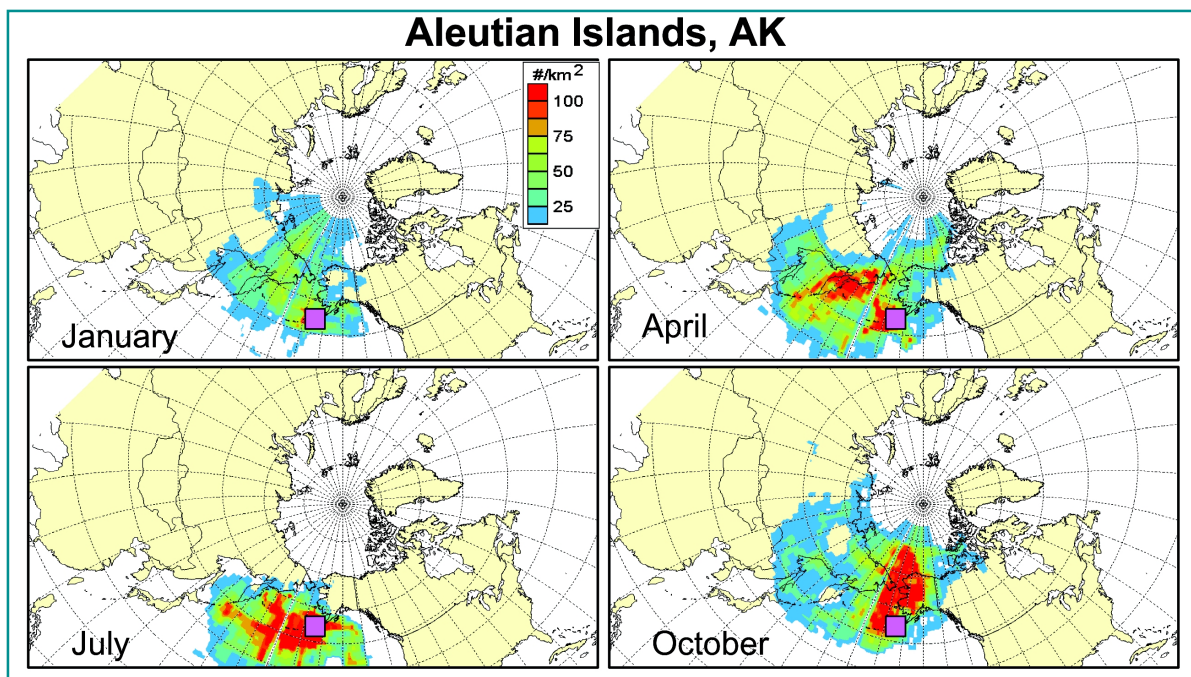

Aleutian Islands, AK

Figure 7-7. Aleutian 10-day back trajectories for the year 1999. Color shading refers to the probability that trajectories passed over a given area before arriving at the receptor site.
Source: Husar and Schichtel (2001).

smoke or dust may not be visible in satellite images, the transport of other airborne compounds, including POPs, still occurs within these pathways.

During the springtime, after the winter snows have melted and before a vegetation cover has appeared, extensive clouds of yellow dust can be raised from the Gobi and Taklamakan Deserts (Duce, 1995). Strong cold fronts originating in Siberia are associated with strong upward air motions in front of them as they travel southward. The vertical motions carry whatever pollutants are found near the surface upward to the altitude of the jet stream, where they can be rapidly transported eastward to North America (Figure 7-3). Siberian cold fronts can travel all the way to southern China, so a broad range of pollutants from different regions can be transported to North America during a single frontal passage. Sinking air over northwestern North America allows suspended material in the air to be brought to the surface. It is only during the spring that the effects of these events are visible in satellite images. The frontal passages occur at other times, and may transport pollutants to North America without raising dust clouds.

Figure 7-8 shows the results of a three-dimensional, chemistry-transport model simulation of the transport of dust from the Gobi Desert to North America during the dust storm of April 1998 (Hanna et al., 2000). The "dust" plume shown in Figure 7-8 was produced by both horizontal and vertical motions. In general, long-range transport of pollutants involves movement vertically as well as horizontally. Events such as those shown in Figure 7-8 result in rapid transport, on the order of a few days, over transcontinental distances. This simulation included emissions from different points within Asia and the effects of atmospheric mixing during the transport of the emissions. Emissions from different regions of Asia follow their own pathways to North America, where they affect different areas. Figure 7-8 also shows that transport does not terminate abruptly at the coast of North America; rather, there can be deposition on mountains near the coast and even further inland. The frequency of these events and their importance as a transport mechanism to North America remain to be determined. The next step will be to include in the model the chemical losses and multimedia transfers described above so that the transmission of POPs and other chemicals during intercontinental transport can be determined.

The results of a three-dimensional, chemistry-transport model simulation of the global distribution of hexachlorobenzene (HCB), a long-lived POP, are shown in Figure 7-9 (Olaguer and Pinto, 2001). The distribution of HCB was calculated using data for the emissions of HCB and loss by OH radicals calculated with the model. The areas of high concentration in Figure 7-9 basically result from a combination of high emissions and meteorological conditions that favor trapping emissions close to the surface. The model was able to simulate successfully a number of observed features of the global distribution of HCB, including its ratio between the Northern and Southern Hemispheres, lending credence to this approach. Simulations
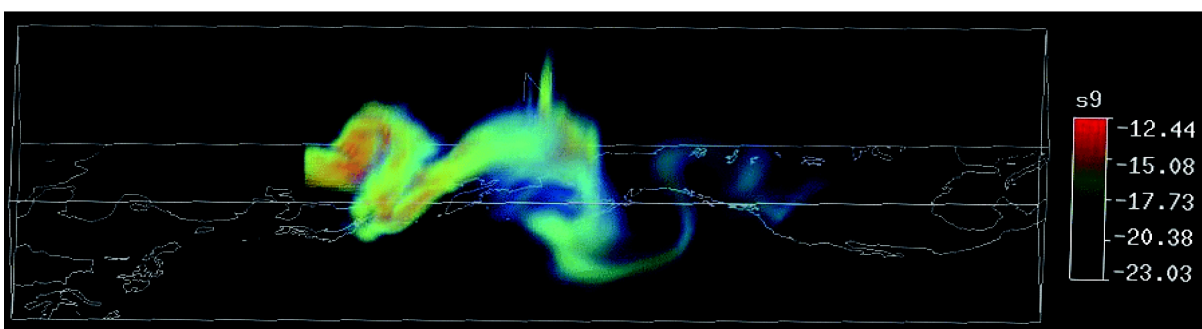

Figure 7-8. Forward trajectory simulation of dust from the Gobi Desert to North America, April 1998.

such as these will permit quantitative testing, by comparison with observations, of many of the concepts presented in this chapter.

POPs Transport in Water

Long-range transport can also occur through hydrologic pathways, with POPs entrained on sediment, in microscopic species, or in solution (for the more water-soluble compounds). POPs released or deposited onto terrestrial areas are transported down rivers to oceans, and then potentially to remote locations through oceanic currents. POPs deposited and accumulated on ice in the Arctic can also be transported into the North Atlantic by ice floes. The contribution of hydrologic transport to global POPs pollution has not been quantified, although it is generally considered to be substantially less than occurs through atmospheric transport of these semivolatile, hydrophobic substances.

Oceanic currents can be wind generated at the ocean surface or result from water temperature and density (thermohaline) differences. Wind generated currents are generally limited to the first 1,000 meters of depth, whereas thermohaline currents can extend down to the deep sea. Oceanic current speeds are highly dependent on location. Movement in the Gulf Stream or Kuroshio currents is rapid (4–6 knots) over thousands of miles, whereas within localized gyres (circling currents) or deep oceanic regions little to no net water transport may be occurring. The major surface currents in the North Atlantic Ocean form a large gyre, or closed clockwise circulation, in which the Gulf Stream current flows northward along the east coast of North America to Cape Hatteras, NC, then travels to the northeast, then southward along the coast of Europe, and finally westward across the tropical Atlantic Ocean in the North Equatorial Current to close the circulation (Figure 7-10).

A similar circulation pattern is found in the North Pacific Ocean, in which the Kuroshio Current flows northward along the coast of Asia to the southern east coast of Japan through the East China Sea.
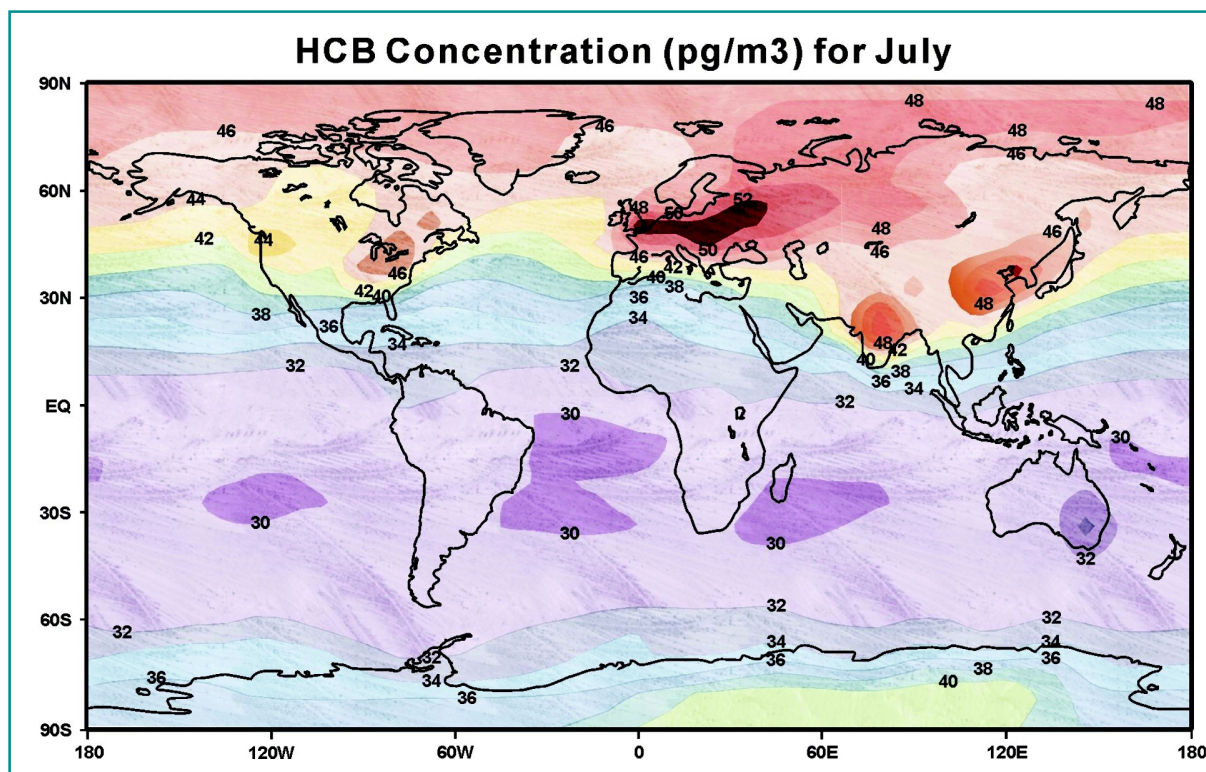

Figure 7-9. Calculated global distribution of hexachlorobenzene at the earth's surface.
Source: Olaguer and Pinto (2001).

Its width is about 100 km, its speed 3–4 knots, and it transports 30–60 million tons of water per second (Pickard, 1975). Pacific currents can transport POPs from Asia to continental Alaska and its island chains. The transit time around the Bering Sea was estimated in the order of 1 year, as measured by satellite tracking of drift markers (Royer and Emery, 1984). Measurements of the latitudinal distribution of POPs in seawater are sparse. Concentrations of at least one organochlorine, α-HCH, in seawater were found to increase with latitude by roughly a factor of 20 on a transect from the Java Sea to the Beaufort Sea (Wania and MacKay, 1996; and references therein). This increase is probably the result of atmospheric deposition. Although computer models of the ocean's circulation exist and have been coupled with atmospheric models to study problems related to climate change, they have not yet been applied to problems of POPs transport.

The ocean can also act as an ultimate sink for POPs, either through the deposition of dead biological organisms or via deep-current circulation. POPs can be concentrated up the marine food chain, starting with phytoplankton. Some POPs are transported to deeper layers of the ocean by settling phytoplankton, or "marine snow." Evidence for this pathway is provided by elevated concentrations of POPs in marine fish, with higher concentrations found in deeper living varieties (Froescheis et al., 2000; Looser et al., 2000). Sinking water, such as part of the Gulf Stream near Iceland, also carries with it pollutants such as POPs. POPs transported to the deep sea by either of these pathways will probably remain there and be degraded, because it takes several hundred years for water in the deep sea to return to the surface again in a region of upwelling.

POPs Transport by Migratory Animals

POPs transport through migratory species is also considered a potential source of contaminant movement under the Stockholm Convention. Evaluation of this mechanism differs from the previous assessments of atmospheric and hydrologic transport, because POPs transported through migratory species are often focused on a localized region (lakes, nesting sites) or injected directly into the food supply. Through this means, the POPs concentration in lipid is maintained, dilution is
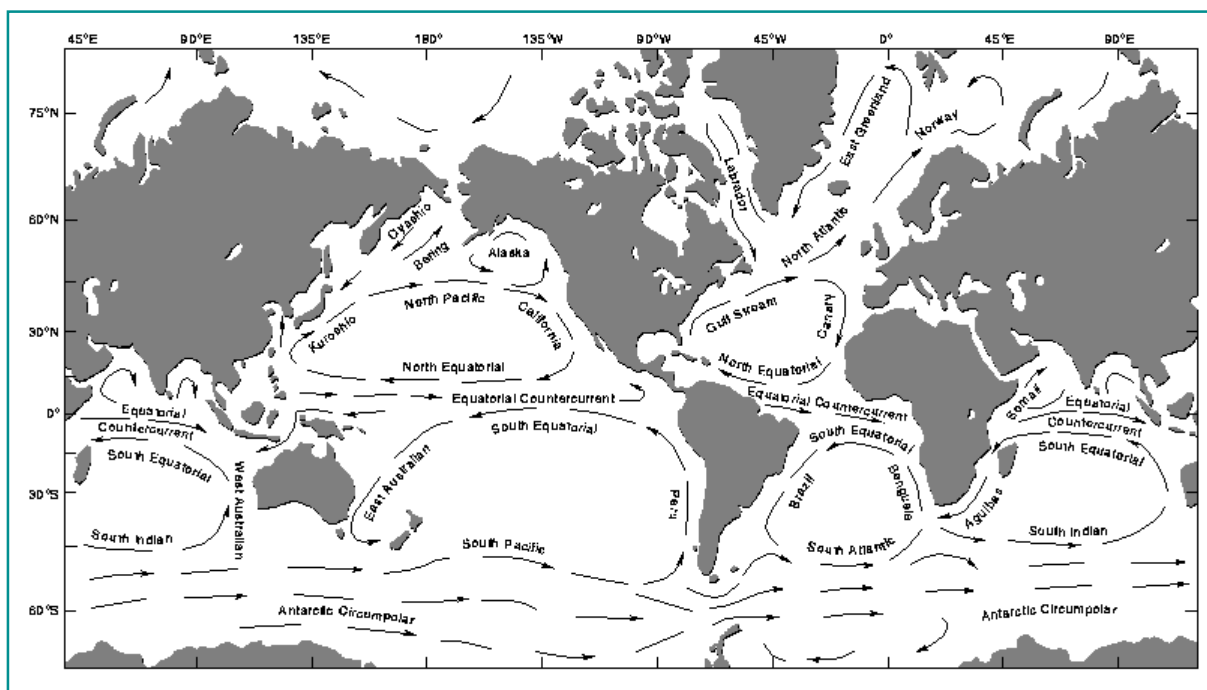

Figure 7-10. Oceanic surface currents.
Source: Adapted with permission from Apel (1987); NOAA.

prevented or minimized, and the POPs are potentially targeted at a receptor animal species. This can be viewed as more of a stiletto approach, compared with the blunderbuss of atmospheric transport.

The bulk amount of POPs transported long distances by migratory birds, fish, and/or marine mammals is highly uncertain, and estimates are not available (van de Meent et al., 2001). However, it has been calculated that the POPs loading from spawning and dying salmon swimming to localized lakes in Alaska is greater than air deposition levels (Ewald et al., 1998). POPs may also be transported by migratory birds to remote rookeries, and from there be transferred to resident species. Most importantly for high trophic predators and human risks, the oceanic movement of some fish and marine mammals can transfer the POPs loads obtained throughout this migratory journey directly to the end predator.

The Future

Atmospheric monitoring and modeling provide reassurance that efforts on POPs can be, and are, effective in regions such as the Great Lakes. On the basis of data collected by the Integrated Atmospheric Deposition Network (IADN; *www.epa.gov/ glnpo/iadn/*), trends in concentrations of a number of key organochlorine toxins can be calculated and extrapolated to give approximate dates when atmospheric concentrations will be beneath minimum detection limits of the measurement techniques used (i.e., "virtual elimination"; Cortes et al., 1998). Results of these extrapolations are shown in Figure 7-11. Atmospheric levels of DDT and DDD are predicted to be the first below detection limits, disappearing at all sites by about 2010. HCB will remain in the atmosphere the longest of all the compounds considered, mainly because of its continued production as a byproduct. Overall, though, these data suggest that most of the compounds will disappear from the atmosphere of the Great Lakes by the middle of this century.
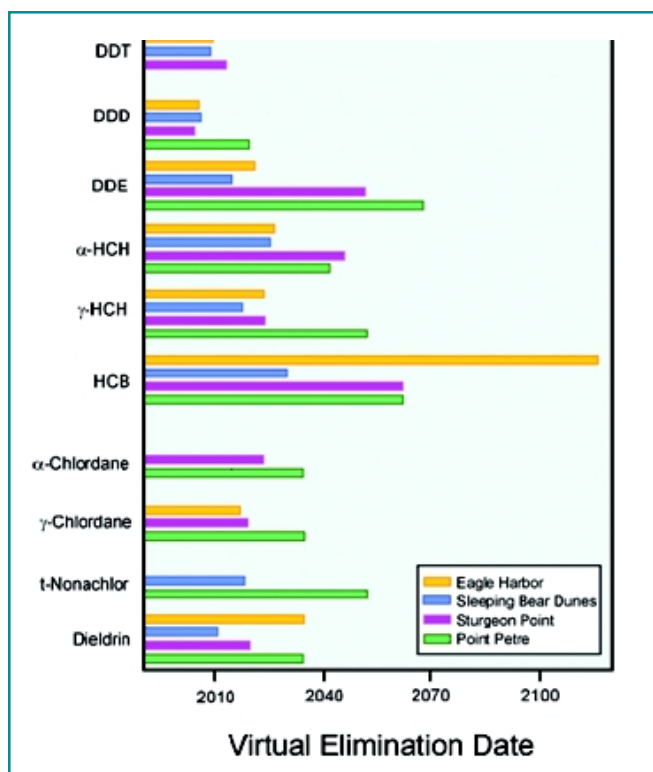

Figure 7-11. IADN virtual elimination dates. Virtual elimination date estimates assume no significant new sources.
Source: Cortes et al. (1998).

Acknowledgments

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